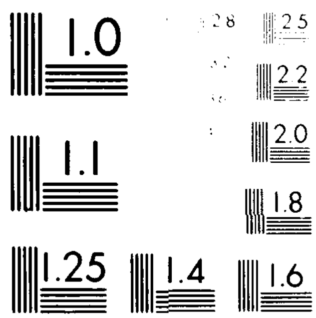
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The view, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The dynamics of high frequency phonons has proven to be a difficult area of investigation. The general problem we have investigated is one of developing optical techniques to study high frequency phonon dynamics. A statement summarizing the problem, as we have examined it, can be most conveniently presented in four parts as described below. (1) The mechanisms of decay and lifetime of high frequency phonons ($\lesssim$ 200 GHz) including the case of strong resonant trapping.

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20. ABSTRACT CONTINUED

(2) Generation of high frequency phonons by stimulated emission under conditions of a population inversion. (3) Development of computer simulation techniques to describe the dynamics of high frequency phonons resonantly trapped by an inhomogeneously broadened resonance with inclusion of stimulated emission. (4) Development of techniques to reduce the spectral bandwidth of a coaxial flashlamp-pumped tunable dye laser for use in problems (1) and (2). Problem (1) and (2) have been examined for several insulating ionic solids incorporating small amounts of impurity ions whose electronic states are accessible with visible light. The electronically excited ions serve to generate, detect and resonantly trap the nearly monoenergetic distribution of high frequency phonons.

Statement of the Problem

The dynamics of high frequency phonons has proven to be a difficult area of investigation. The general problem we have investigated is one of developing optical techniques to study high frequency phonon dynamics. A statement summarizing the problem, as we have examined it, can be most conveniently presented in four parts as described below.

(1) The mechanisms of decay and lifetime of high frequency phonons ($\geq$ 200GHz) including the case of strong resonant trapping.

(2) Generation of high frequency phonons by stimulated emission under conditions of a population inversion.

(3) Development of computer simulation techniques to describe the dynamics of high frequency phonons resonantly trapped by an inhomogeneously broadened resonance with inclusion of stimulated emission.

(4) Development of techniques to reduce the spectral bandwidth of a coaxial flashlamp-pumped tunable dye laser for use in problems (1) and (2).

Problems (1) and (2) have been examined for several insulating ionic solids incorporating small amounts of impurity ions whose electronic states are accessible with visible light. The electronically excited ions serve to generate, detect and resonantly trap the nearly monoenergetic distribution of high frequency phon... Problem (1) has been investigated in $Al_2O_3:Cr^{3+}$ (ruby) at 0.87THz, in $LaF_3:Pr^{3+}$ at 0.77THz, and in CaF_2Sm^{2+} at 3.87 THz while problem (2) has been studied for $LaF_3:Er^{3+}$ at ~0.2THz.

Summary of Most Important Results

Optical techniques for generation and detection of high frequency phonons provide a powerful means to study the dynamics of high frequency phonons. The phonons are resonantly trapped so that losses due to fundamental decay processes can be extracted. Results so far indicate anharmonic decay rates which are generally but not always, consistent (order of magnitude) with theoretical estimates based on an isotropic Debye solid, but we have not yet been able to perform systematic studies of the frequency dependence of the decay. Resonant phonon assisted energy transfer has been identified for the first time and has been related to experiments on energy transfer in ruby. Stimulated emission of tunable high frequency phonons, with high gain, has been demonstrated under conditions in which the population inversion can be maintained for times sufficiently long for phonon propagation over large distances ($\sim 1 cm^{-1}$), offering the possibility of coherent phonon generation.

Described below are a few of the more important details about our major results.

(1) Decay of High Frequency Phonons

(a) LaF_3Pr^{3+} (Publication 1) - The 0.77 THz phonons are strongly trapped in the volume containing the excited Pr^{3+} ions and decay in 20 ns predominantly by anharmonic breakup (3-phonon) processes of the longitudinal phonons.

(b) $CaF_2:Sm^{2+}$ - The 3.87 THz phonons were not resonantly trapped so that their lifetime was limited by spatial diffusion from the optically excited volume. A lower limit of 50 ns was established however for the lifetime of the longitudinal phonons at this frequency. This is much longer than would be predicted by an isotropic Debye model of anharmonic decay.

(c) $Al_2O_3:Cr^{3+}$ (Publications 3 and 6) - The decay of 0.87 THz phonons was studied as a function of excited ion concentration (resonant scatterers), size of resonant scattering volume, and concentration of Cr^{3+} ions. The decay showed evidence for spatial diffusion for small volumes or low excited ion densities. Under conditions of strong scattering we have observed, for the first time, resonant phonon-assisted energy transfer (RPAET) whereby a phonon initiates energy transfer between an excited Cr^{3+} ion and a ground state Cr^{3+} ion, with a shift in phonon frequency making up the energy mismatch between the excited states of the two Cr^{3+} ions due to inhomogenieties in the local strain. A model was developed which was consistent with the absolute magnitude of the observed phonon loss rates ($\approx 10^6\ s^{-1}$) as well as its dependence on the variables in the experiment (concentration, spot size, laser power, etc.). The results require a Cr^{3+}-Cr^{3+} exchange interaction which is an order of magnitude less than had been previously estimated and the model is consistent with the observed absence of RPAET as an energy transfer mechanism in fluorescence line narrowing experiments and the failure to detect long range energy transport in transient grating experiments. We can also place a limit of 1 μs on the lifetime of 0.87 THz longitudinal phonons in ruby, an order of magnitude greater than theoretical estimates suggesting that in real materials high frequency phonon lifetimes can be quite long.

(2) Stimulated Emission of Phonons (Publications 7 and 8) - Stimulated emission of phonons in the 0.2 THz frequency range was observed in $LaF_3:Er^{3+}$. The phonons were generated in the relaxation between the Kramers components of the $^4S_{3/2}(1)$ state of Er^{3+} split in a magnetic field after inverting the population with a pulsed

tunable dye laser. The stimulated emission was identified from the temporal behavior of the luminescence from the upper Kramers component which showed a speedup in the relaxation during inversion. The population dynamics were analyzed with the rate equations for the system. Comparison with experiment indicated strong preferential stimulated emission along the axis of the cylindrically excited volume and a gain of up to 5 mm^{-1}. The population inversion existed sufficiently long for phonon propagation over macroscopic distances and the phonon frequency was tunable with the magnetic field. The parameters obtained from the analysis of the experiment indicate that coherent generation in a cavity formed by the crystal surfaces may be possible.

(3) -Computer simulation of phonon dynamics - Developmental work was carried out to use computer simulation techniques to model the phonon dynamics. The program has progressed to the point where phonons can be created at any point in a 3-dimensional excitation cylinder with random walk scattering until the phonon escapes through the boundaries of the excited volume. The phonon loss exhibits three regimes: early time where phonons are lost only infrequently, medium time where the number of phonons drops off rapidly (nearly exponentially), and long time where a slowly decreasing tail signals extensive scattering within the excitation volume.

(4) Techniques to spectrally narrow a flashlamp-pumped tunable dye laser (Publication 5) - Several techniques to spectrally narrow the output of a flashlamp-pumped tunable dye laser were successfully developed for use in our phonon experiments.

(a) Grazing incidence feedback from a grating mirror combination yielding with R6G 50 mJ pulses with a 0.2 cm^{-1} bandwidth,

an output energy comparable with that obtained using a grating in Littrow but with one tenth the bandwidth.

(b) Injection locking - 20 mJ 0.1 cm^{-1} bandwidth, 100 ns pulses were obtained by cavity dumping after injection of the output of a N_2-pumped dye laser. A Pockels cell was utilized to switch the cavity gain.

(c) Alternating high gain-low gain cavities - 30 mJ, 0.2 cm^{-1} pulses were obtained with the lower gain coumerin dyes by alternating the photons between the low feedback grazing incidence grating and a high feedback mirror using a beam splitting polarizer and wave retarder. This raised output energies by a factor of 5 over the energies available using only the grating-mirror combination.

All of these techniques have proved useful. Methods (a) and (c) produced the most energy, but (b) was capable of producing variable pulse widths.

List of Publications

1. "Anomalous Luminescence Decay in $LaF_3:Pr^{3+}$ Due to Resonant Trapping and Bottlenecking of 23 cm^{-1} Phonons," L. Godfrey, J. E. Rives, and R. S. Meltzer, J. Lumin. 18/19, 929(1979).
2. "Quadraxial High Repetition Rate Adaptor for Coaxial Flashlamp Pumping of Laser Dyes," L. A. Godfrey, R. S. Meltzer, and J. E. Rives, Appl. Opt. 18, 602(1979).
3. "Dynamics of 29 cm^{-1} Phonons in Ruby Using Optical Detection," W. C. Egbert, R. S. Meltzer and John E. Rives, Phonon Scattering in Condensed Matter, (ed. H. J. Maris, Plenum, New York, 1980) 365.
4. "Exciton Dynamics Within an Inhomogeneous Line," H. T. Chen and R. S. Meltzer, Phys. Rev. Letters 44, 599(1980).
5. "Grazing Incidence Grating Techniques for Flashlamp-Pumped Tunable Dye Lasers," L. A. Godfrey, W. C. Egbert, and R. S. Meltzer, Opt. Commun. 34, 108(1980).
6. "Resonant Phonon Assisted Energy Transfer in Ruby from 29 cm^{-1} Phonon Dynamics," R. S. Meltzer, J. E. Rives, and W. C. Egbert, Phys. Rev. (1982) in press.
7. "Stimulated Emission of Tunable High Frequency Phonons in LaF_3Er^{3+}," D. J. Sox, J. E. Rives, R. S. Meltzer, Journal de Physique, Colloque C6-1981, 450 (1981).
8. "Stimulated Emission of 0.2 THz Phonons in $LaF_3:Er^{3+}$," D. J. Sox, J. E. Rives and R. S. Meltzer, Phys. Rev. (1982), in press.

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1. "Optical Studies of the Dynamics of 116 cm^{-1} Phonons in $CaF_2:Sm^{2+}$," W. C. Egbert, J. E. Rives, and R. S. Meltzer, Second Conference on Dynamical Processes in the Excited States of Ions and Molecules in Solids, Madison, Wisconsin, June 18-20, 1979, Bull. Am. Phys. Soc. 24, 897(1979).
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6. "Stimulated Emission of High Frequency Phonons in $LaF_3:Er^{3+}$," J. E. Rives, R. S. Meltzer, and D. J. Sox, Dynamical Processes Conference, Regensburg, West Germany, July 28-31, 1981, Bull. Am. Phys. Soc. 27, 67(1982).

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